Pinned Spin Depth Profile of an Oxidized-Mn/$Ga_{1-x}Mn_xAs$ Exchange Bias Bilayer – The Effects of Overannealing

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Abstract— Mn/$Ga_{1-x}Mn_xAs$ bilayers can exhibit large exchange bias if annealed for an optimal period of time to transform the Mn cap into antiferromagnetic MnO. Annealing for too long tends to drastically increase the number of spins that become pinned along the direction of a cooling field, but it also results in samples that exhibit little or no exchange bias. To investigate why the increase in pinned spins does not enhance the exchange coupling, we used polarized neutron reflectometry to examine the magnetic depth profile of a typical overannealed Mn/$Ga_{1-x}Mn_xAs$ bilayer. We observe a large magnetization with a pinnable component *distributed throughout the cap*. This shows that overannealing results in a cap that is not entirely antiferromagnetic, and that the additional pinned spins do not originate from uncompensated moments directly at the antiferromagnet/ferromagnet interface – explaining why they do not contribute to the exchange bias coupling.

***Index Terms*—Exchange Bias, Magnetic Semiconductors, Neutron Scattering.**

I. Introduction

EXCHANGE BIAS (H_E) is the shift of the ferromagnetic hysteresis loop about the zero of applied field (H), and results from exchange coupling between unpinned spins and pinned spins (i.e. spins with a very large coercive field relative to the rest of the system).[1,2,3,4,5]. For the case of ferromagnetic exchange coupling between an antiferromagnet (AFM) and a ferromagnet (FM), the pinned spins are at or near the AFM/FM interface, and align parallel to the cooling field H_{Cool}. Such pinned spins can sometimes be detected as a small vertical shift of the magnetization (M) in the hysteresis loop[6]. Recently, exchange bias has been observed in $Ga_{1-x}Mn_xAs$ films [7,8] capped with antiferromagnetic (AFM) MnO, the first example of an exchange biased dilute FM semiconductor.[9,10,11] In such systems, the MnO is formed by annealing a Mn-capped $Ga_{1-x}Mn_xAs$ film in air. We find that variations in annealing time for a given growth drastically affect the resultant magnetic properties of Mn-capped $Ga_{1-x}Mn_xAs$ samples[12]. If the sample is not annealed, no H_E or pinned M is observed, consistent with ref. 9 & 10. For optimal annealing times (which we find to be different from growth to growth), the largest H_E is observed, with little or no pinned M. However, if the sample is annealed for too long, a pinned M along the H_{Cool} direction becomes progressively *larger* (implying an increased number of pinned spins), while H_E becomes progressively *smaller*. This result is counterintuitive, since pinned spins are necessary for exchange bias.

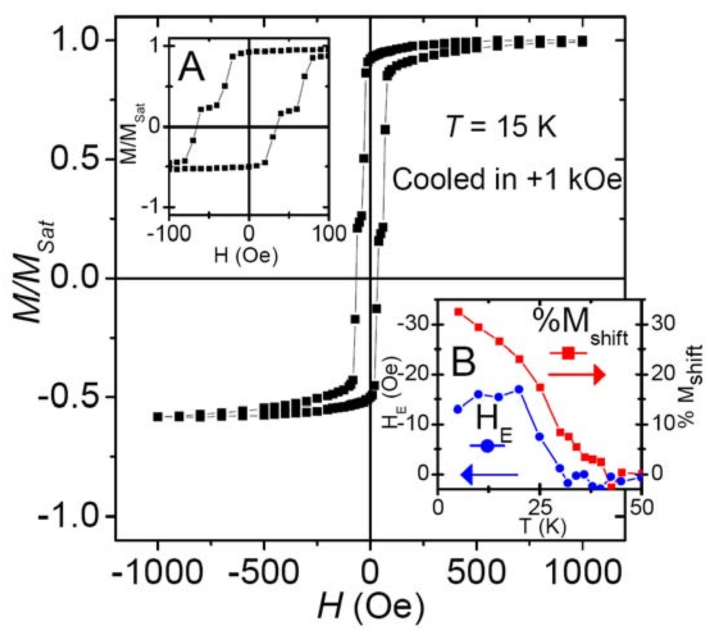

Fig. 1. SQUID magnetometry results for an overannealed, oxidized Mn / $Ga_{1-x}Mn_xAs$ bilayer. Inset A zooms in to show the small H_E. Inset B shows the temperature dependencies of H_E (open circles, left scale), and % magnetization shift (closed squares, right scale).

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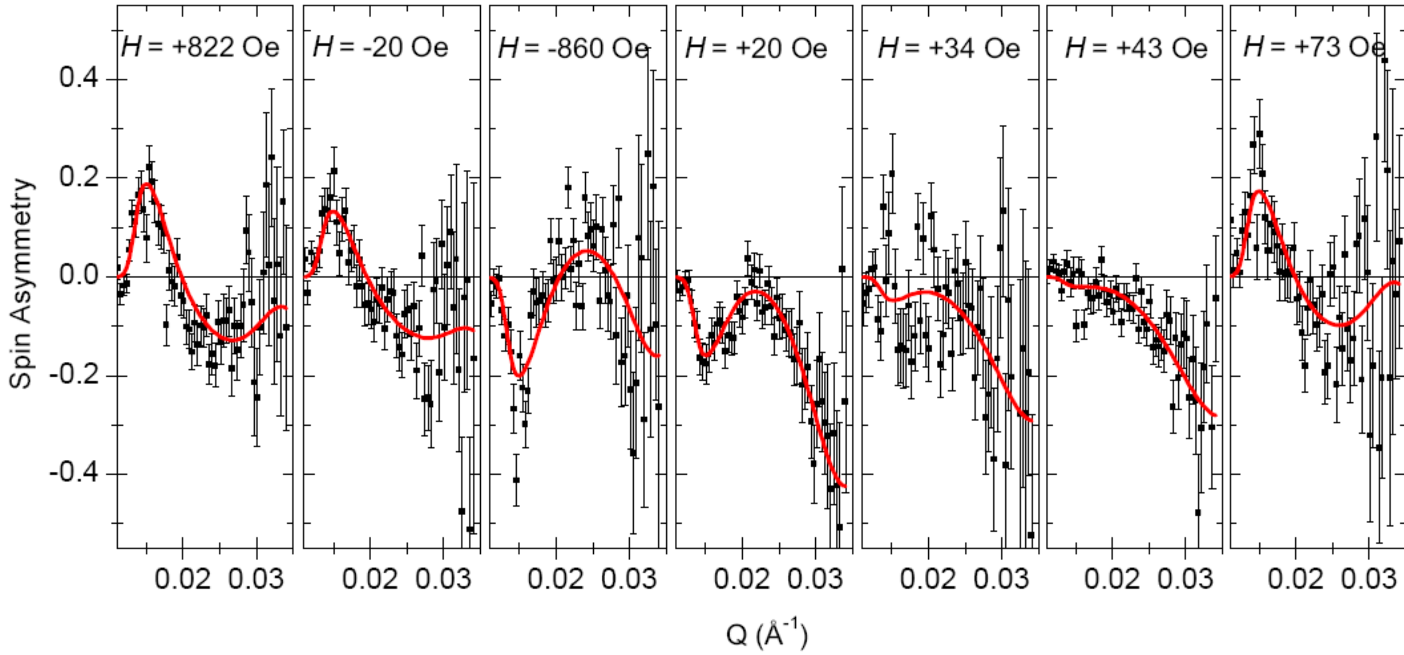

Fig. 2. Field dependent PNR data (symbols) and fits (lines) expressed as spin asymmetry. Note that the +822 Oe data and the -860 Oe data are *not* totally anti-symmetric, as would be the case if the magnetization depth profiles at those fields were perfectly anti-symmetric.

II. EXPERIMENTAL RESULTS

The sample we consider is a 30 nm $Ga_{0.94}Mn_{0.06}As$ film grown on a (001) GaAs substrate, and capped with about 10 nm of oxidized Mn. The sample was produced via molecular beam epitaxy[11,13] using a non-bonded sample holder, and was annealed in air for 1 minute at 130 ºC. Fig. 1, shows results from SQUID[14] magnetometry along the [100] direction (easy-axis) of the sample after applying H_{Cool} = 1 kOe[15]. The main part of Fig. 1 reveals an exceptionally large M shift indicating a large number of pinned spins.[16] Inset A zooms in to show a small H_E = -15 Oe, and steps indicative of two decoupled magnetic phases. Inset B shows the temperature dependencies of % M shift[17] and H_E. The large M shift and small H_E indicate that the annealing was not optimal, and imply that the sample was *over*annealed.

Despite having a larger number of pinned spins, H_E for this sample is below values previously reported by our group for similar samples with no M shift. To understand why this is so, we used polarized neutron reflectometry (PNR) to determine the spatial location of the pinned spins. In a PNR measurement, the chemical scattering length density (ρ_{Chem}, related to the chemical composition) and M depth profiles of the sample can be inferred from the spin-dependent reflectivities[18,19,20]. Using the Asterix polarized neutron reflectometer/diffractometer at Los Alamos National Laboratory we performed such measurements as a function of H along the [100] of the above described sample, after applying H_{Cool}. We use the convention that the direction of H_{Cool} be defined as *positive H*, and all other properties (spin-up, spin-down, etc.) are defined relative to that.[21]

Fig. 2 shows the measured reflectivities and fits expressed as spin asymmetry (the difference in the spin-up reflectivity and the spin-down reflectivity divided by the sum), as H is cycled through the hysteresis loop. Spin asymmetry is particularly sensitive to the projection of M along H, making it a useful representation for highlighting magnetic features in PNR data from low-M systems like $Ga_{1-x}Mn_xAs$.[22] Note that at the two field extremes (H = +822 Oe and H = -860 Oe, which Fig. 1 shows to be the positive and negative "saturation" states[23]), the respective reflectivities are *not* completely anti-symmetric (particularly evident at $Q \approx 0.025$ Å^{-1}) – as would be the case if the respective M profiles were equal in magnitude, but opposite in sign.

The models used to fit the Fig. 2 data are shown in Fig. 3[24]. An additional set of PNR data were taken out to higher Q (0.057 Å^{-1}) than the sets shown in Fig. 2, and were used to determine the ρ_{Chem} profile (panel A, Fig. 3) for all of the field states. Panel A shows the GaAs substrate, the $Ga_{1-x}Mn_xAs$ film, and a very rough (or chemically nonuniform) oxidized Mn cap all clearly delineated from one another. Panels B-H contain the M profiles corresponding to the data in Fig. 2. In addition to the expected M of the $Ga_{1-x}Mn_xAs$ layer, the *cap* is also magnetized throughout its thickness. While optimal annealing in air presumably changes the cap from Mn to MnO, our data indicate that *over*annealing further changes the cap composition, creating a magnetic compound in addition to (or in lieu of) the desired AFM MnO. The cap saturation M is comparable in magnitude to that of the $Ga_{1-x}Mn_xAs$ layer, but responds to field much differently – identifying the origin of the two decoupled magnetic phases suggested by SQUID.

Panel I shows the addition of profiles for the two saturation states (panels B and D) divided by two. The result is the portion of M that remains pointed along the direction of H_{Cool} even after applying large fields in opposite directions – i.e. it is the pinned M depth profile. Panel I shows that the large M shift in Fig. 1 originates from pinned spins distributed

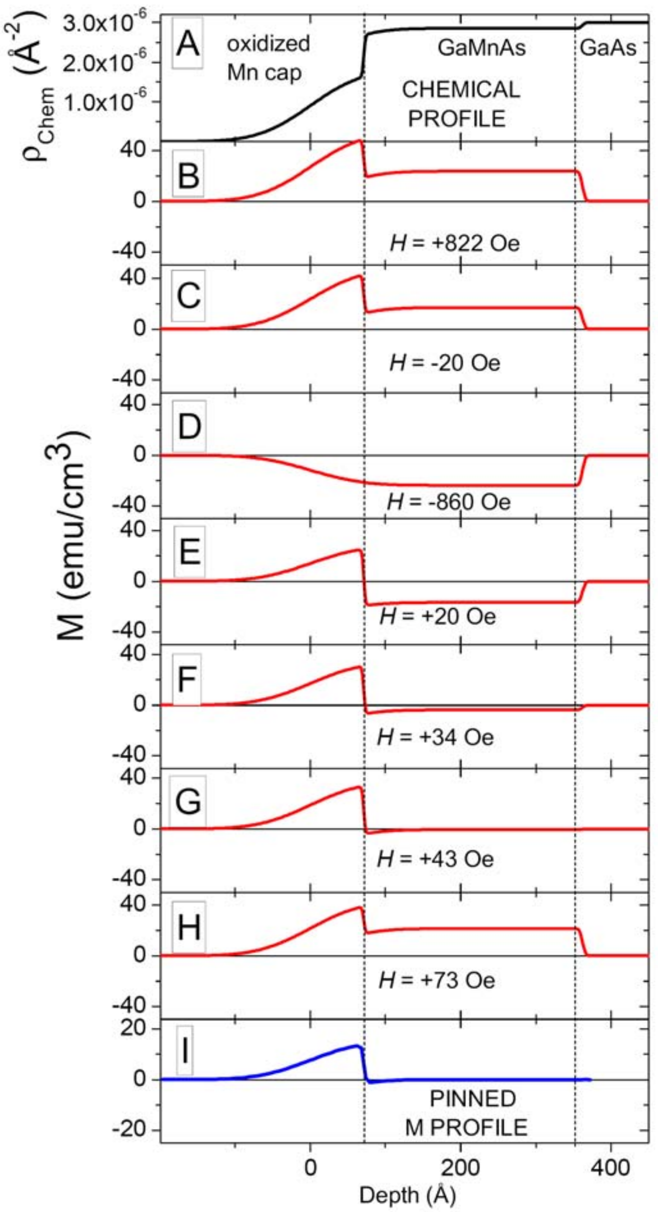

Fig. 3. Depth profiles determined from fits to the data in Fig. 2. Panel A shows the chemical scattering length density, Panels B-H show the field dependent magnetization, and panel I shows the pinned magnetization. All detectable pinned spins are localized in the overlayer.

throughout the oxidized Mn cap, as approximately 25% of the total cap M is pinned along the H_{Cool} direction. Comparison of the integrated B and D profiles indicates a +11% M shift, in reasonable qualitative agreement with Fig. 1[25].

III. CONCLUSIONS

Optimal annealing of a Mn/$Ga_{1-x}Mn_xAs$ bilayer in air produces an AFM MnO capping layer that can exchange couple to the FM $Ga_{1-x}Mn_xAs$ layer [9,10], presumably via a very small number of interfacial pinned spins that have so far eluded detection. SQUID shows that annealing for an excess period of time greatly increases the number of spins that can become pinned along H_{Cool}, but it does not strengthen the exchange coupling. PNR reveals that this is because the overannealed cap is not AFM (on average), and that the additional pinned spins are distributed *throughout* the cap. So, the additional pinned spins do not arise from uncompensated moments within a few nm of an AFM/FM interface, and therefore do not contribute to the AFM/FM exchange coupling thought to be present in optimally annealed samples. Despite the transformation of the cap, a small H_E persists, possibly due to AFM/FM exchange coupling of interfacial remnants of AFM MnO with $Ga_{1-x}Mn_xAs$, weak exchange coupling between the bulk of the cap and the $Ga_{1-x}Mn_xAs$, or exchange coupling between the pinned and unpinned components of the cap.

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[14] Superconducting QUantum Interference Device
[15] 1 kOe = $4\pi A{\cdot}m^{-1}$
[16] Similar measurements with opposite H_{Cool} showed that the sign of the M shift is correlated with the sign of H_{Cool}, proving that the shift is not due to an instrumental bias.
[17] Defined as the percentage difference between the absolute values of M(+1 kOe) and M(-1 kOe).
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[21] The polarizing optics of the reflectometer define a positive field direction, and the sign of H must be positive to maintain a polarized beam. Measurements after applying H_{Cool} = +822 Oe got the positive H side of the hysteresis loop, and measurements after applying H_{Cool} = -860 Oe to got the negative side.
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[23] Saturation, as used here, refers to the field at which $M(H)$ becomes constant. Presumably, the pinned M becomes *un*pinned at high enough fields.
[24] 1 $emu{\circ}cm^{-3} = 10^{-3}\ A{\cdot}m^{-1}$.
[25] The quantitative disagreement in this value between SQUID and PNR could be due to uncertainty in sample temperature for each measurement.